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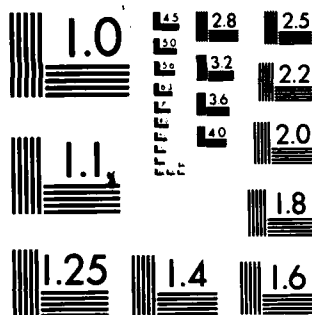
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ELECTROCHEMICAL REDUCTION REACTIONS INVOLVING FORMIC ACID

by

M. H. Miles, A. N. Fletcher, and G. E. McManis

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) Various investigators have shown that the electrode reduction of CO_2 in water using metal electrodes yields acid as the main product. Recent publications have generated conflicting claims regarding the further reduction of formic acid to methanol. Our studies using platinum electrodes at a fixed pH in mildly acidic NaClO_4 solutions show an increase in the cathodic current when NaCOOH is added. Closer examinations show that the $\text{HCOOH}/\text{HCOO}^-$ equilibrium is involved and that H_3O^+ rather than HCOOH is reduced. The results of these		

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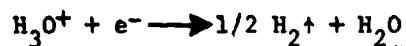
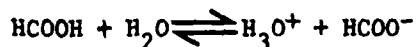
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Investigations can be represented by the reaction sequence



where the net result is the reduction of protons present as either undissociated HCOOH or H_3O^+ to form hydrogen gas. The positively charged protons complexed with one or more water molecules are electrochemically reduced at a more positive potential than neutral water molecules.

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ELECTROCHEMICAL REDUCTION REACTIONS INVOLVING FORMIC ACID

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INTRODUCTION

Electrochemical reduction provides a means of activating carbon dioxide for the production of fuels and organic chemicals. The electrode reduction of CO_2 in water using metal electrodes yields formic acid and formate ions as the main products [1-7], i.e., $\text{CO}_2 + \text{H}_2\text{O} + 2\text{e}^- \rightarrow \text{HCO}_2^- + \text{OH}^-$. Previous studies have shown that the further reduction of formic acid to methanol at metal electrodes occurs only in a narrow potential range and at impractically small current densities [2,7]. Cyclic voltammograms reported for TiO_2 electrodes in N_2 -saturated KCl solutions show increasing cathodic currents with increasing concentrations of formic acid [8]. Furthermore, the direct reduction of CO_2 to methanol is reported for TiO_2 -Ru cathodes [9]. Augustynski and co-workers [8] claim that the reduction of formic acid takes place at potentials positive to hydrogen evolution on TiO_2 electrodes. This conflicts with conclusions by Tinnemans et al. [10] who claim that neither formic acid nor formate ion is the electroactive species being reduced.

These authors propose instead that the cathodic currents observed result from local pH changes at the TiO_2 /solution interface and involve the buffering action of the formic acid/formate system [10]. This explanation has been rejected by Augustynski [11].

Our potential scan experiments in NaClO_4 solutions at a constant pH have revealed that the addition of sodium formate under mildly acidic conditions increases the cathodic current on a variety of metal electrodes. Detailed studies are reported here for experiments conducted using platinum electrodes.

EXPERIMENTAL SECTION

Solutions (1.0 M) were prepared by dissolving 7.02 g $\text{NaClO}_4 \cdot \text{H}_2\text{O}$ in 50 ml of deionized water. The desired formic acid/formate concentrations were produced by additions of NaCOOH and HClO_4 . The platinum wire working electrode ($d = 0.1$ cm, $l = 0.7$ cm, $A = 0.22$ cm²) was constructed by using heat shrink Teflon to seal the wire in glass tubing. Exhaustive electrolysis experiments used a large platinum sheet electrode ($A = 50$ cm²). The platinum counter electrode was isolated from the main compartment of the beaker-type cell by a section of glass tubing with an ultra-fine glass frit at the bottom. All potentials were measured against a saturated calomel electrode (SCE). Electrochemical measurements were always made in helium-saturated solutions.

The solution pH was monitored using the palladium-hydrogen (Pd-H) electrode prepared as described by Gileadi [12]. This electrode was calibrated by measuring its potential versus SCE in standard buffer solutions (Van-Lab, pH 4, 7 and 10). The experimental relationship of pH and potential (E) of the Pd-H electrode versus SCE at 23°C was determined to be

$$\text{pH} = -(E + 0.2031)/0.0575 \quad (1)$$

Adjustments of the pH were generally made by adding measured amounts of 0.50 M HClO_4 to the solutions. Occasionally, the pH was adjusted upward by the use of NaOH solutions.

Cyclic voltammetric studies involved the use of a potentiostat/galvanostat, current-to-voltage converter, and programmer (PAR Models 173, 176, and 175) in conjunction with an X-Y recorder (Hewlett-Packard 7047 A). A strip chart recorder (HP 7100 B) was also used in exhaustive electrolysis studies. The Pd-H versus SCE potentials were measured with both the PAR potentiostat and a digital multimeter (Fluke 8040 A). The continuous recording of the Pd-H versus SCE potential required a high impedance millivoltmeter (Lazar Digital pH) connected in series with the strip chart recorder.

RESULTS AND DISCUSSION

Potential scan experiments at a fixed pH using various metal electrodes in mildly acidic NaClO_4 solutions revealed that there is an increase in the cathodic current when NaCOOH is added. A detailed study of this effect on a platinum electrode at $\text{pH} = 3.4$ is shown in Fig. 1. This increase in cathodic current clearly occurs at potentials positive to the hydrogen evolution wave that begins near -0.9 V versus SCE at this pH. The excellent correlation between the peak current and the concentration of added NaCOOH could easily lead to the erroneous conclusion that formic acid is being reduced. It is important to note that Fig. 1 shows a small reduction wave at $\text{pH} = 3.4$ that begins at about -0.5 V even in the absence of any added formate.

The cyclic voltammograms in Fig. 2 show that a new reduction peak can be produced simply by the addition of a small amount of HClO_4 to the NaClO_4 solution (solid line, $\text{pH} = 2.21$). In the near neutral solution (dotted line, $\text{pH} = 6.7$), no evidence of any reduction peak can be detected; the only wave is reduction of the solvent (H_2O) at the negative limit of the potential scan. The addition of NaCOOH to the acidified solution involves the $\text{HCOOH}/\text{HCOO}^-$ equilibrium and always yields an increase in pH and a decrease in the reduction peak. It is the addition of further HClO_4 required to restore the original pH value that is responsible for the increase in the peak current in NaCOOH solutions

(dashed line, $\text{pH} = 2.22$). This suggests that the electrode reaction is the reduction of the added hydrogen ions that occurs at a less negative potential than the usual hydrogen evolution reaction involving the solvent.

The voltammograms at $\text{pH} = 2.2$ for HClO_4 and for $\text{HClO}_4 + \text{NaCOOH}$ are very similar (Fig. 2), and both show an anodic peak near -0.3 V that could be explained by the oxidation of adsorbed hydrogen produced by the cathodic reaction. The beginning of formic acid oxidation can be seen in Fig. 2 near the end of the anodic sweep for the solution containing NaCOOH . Potential scan experiments in NaClO_4 solutions without any formate present (Fig. 3) show an excellent correlation between the peak current and either the concentration or the activity of hydrogen ions in the solution. This supports the concept that the reduction peaks observed in $\text{HClO}_4 + \text{NaCOOH}$ solutions (Figs. 1 and 2) result from the reduction of hydrogen ions rather than formic acid molecules.

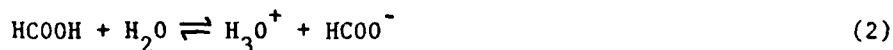
Exhaustive electrolysis experiments using constant current and large electrodes in rapidly stirred solutions yield characteristic inflections in potential as shown in Fig. 4. The time period required for these inflections is mainly dependent on the amount of HClO_4 added to the solution. With equal amounts of added HClO_4 (0.10 ml of 0.50 M HClO_4), the addition of NaCOOH actually decreased the inflection time (broken line, Fig. 4) due to the $\text{HCOOH}/\text{HCOO}^-$ equilibrium that decreased the bulk concentration of hydrogen ions. If experiments are run at the

same initial pH, the presence of NaCOOH yields much longer inflection times than those shown in Fig. 4; these longer inflection times correspond to the additional HClO_4 required to adjust the solution back to the initial pH. The inflections shown in Fig. 4 correspond to $0.89 \text{ e}^-/\text{HClO}_4$ for $\text{NaClO}_4 + \text{HClO}_4$ (solid line) and $0.63 \text{ e}^-/\text{HClO}_4$ for $\text{NaClO}_4 + \text{HClO}_4 + \text{NaCOOH}$. These values are not very reproducible since they depend on the solution stirring rate.

The simultaneous measurement of the solution pH using the Pd-H electrode during constant current exhaustive electrolysis gave pH inflections near $\text{pH} = 7$ as illustrated in Fig. 5. The potential and pH measurements shown in Figs. 4 and 5 involve the same experiments. The shape of the pH curves are typical of those found in textbooks for the titrations of strong and weak acids with a strong base. The pH inflection always came later than the inflection in potential of the working electrode and yielded nearly $1.0 \text{ e}^-/\text{HClO}_4$ (1.04 and $1.01 \text{ e}^-/\text{HClO}_4$ for the two curves shown in Fig. 5). The rate of transport of H_3O^+ to the working electrode becomes the limiting factor before $\text{pH} = 7$ is reached; hence, the inflection comes earlier for this electrode (Fig. 4). This is particularly true for the $\text{HClO}_4 + \text{NaCOOH}$ solution due to both the smaller H_3O^+ concentration and the slower diffusion rate of HCOOH compared to H_3O^+ .

The ultra-fine frit minimizes errors due to the diffusion of hydrogen ions generated at the counter electrode into the main cell compartment. However, after several experimental runs, the counter electrode compartment becomes very acidic ($\text{pH} \sim 0$) and lengthened inflection times ($1.2 \text{ e}^-/\text{HClO}_4$) suggest that extraneous hydrogen ions enter the main cell compartment in measurable amounts. This error becomes negligible if the counter electrode solution is replaced after each run.

The results of these investigations are in accord with the reaction sequence



where H_3O^+ represents the hydrated proton in aqueous solutions. The net result is that the acidic protons present as either undissociated HCOOH or H_3O^+ are electrochemically reduced to form hydrogen gas. The reactive species that is reduced is H_3O^+ rather than HCOOH . Despite being electrochemically inactive towards reduction, the formic acid molecules, nevertheless, serve as a conveyor of protons to the electrode surface. The positively charged proton complexed with one or more water molecules is electrochemically reduced at a more positive potential than neutral water molecules. Preliminary studies on indium electrodes show results similar to those obtained with platinum electrodes; however, the potentials for the cathodic reactions are considerably more negative due to the higher overvoltages for the H_2O and H_3O^+ reductions on indium electrodes [13,14].

The increasing cathodic currents with increasing concentrations of formic acid (0.2×10^{-3} to 8.0×10^{-3} M) reported by Augustynski, et al., [8,11] for TiO_2 electrodes in N_2 -saturated KCl solutions are readily explained by Eqs. (2) and (3). The increase in current reported is due to the reduction of H_3O^+ introduced into the solution by the formic acid additions.

Our studies support the conclusions of Tinnemans, et al., [10] that neither formic acid nor formate ion is the electroactive species being reduced. The experimental data presented by Tinnemans [10] is also in harmony with Eqs. (2) and (3). Obviously, surface pH and buffer capacity are important factors in an electrode reaction that involves the reduction of H_3O^+ .

These results indicate that formic acid molecules cannot effectively compete with H_3O^+ for reduction in acidified solutions. This is especially true at potentials negative to the zero-charge potential where the surface concentration of positively charged species is enhanced [5]. The electrode reduction of formic acid or formate ions to methanol at practical current densities will probably require alkaline solutions to suppress the H_3O^+ reduction.

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FIGURE CAPTIONS

Fig. 1. Potential scan experiments at 20 mV/s in 1.0 M NaClO_4 solutions at pH = 3.4 and 22°C. Concentrations of added NaCOOH were 0, 1.5, 6.0, 13, and 19.5 mM. Geometrical area of the Pt wire electrode was 0.22 cm².

Fig. 2. Cyclic voltammograms at 20 mV/s in 1.0 M NaClO_4 solutions at 22°C. The dotted line is for the solution before HClO_4 or NaCOOH were added, pH = 6.7. The solid line is for the solution containing added HClO_4 (0.10 ml of 0.50 M), pH = 2.21. The dashed line is for the solution containing both NaCOOH (0.0084 g, 2.5 mM) and HClO_4 (0.25 ml of 0.50 M), pH = 2.22. Geometrical area of the Pt wire electrode was 0.22 cm².

Fig. 3. Potential scan experiments at 20 mV/s in 1.0 M NaClO_4 solutions at 22°C containing HClO_4 additions that yield pH = 3.13, pH = 2.22 and pH = 1.95. The volumes of 0.50 M HClO_4 added were 0.01 ml, 0.10 ml, and 0.20 ml, respectively. Geometrical area of the Pt wire electrode was 0.22 cm².

Fig. 4. Potential-time traces for exhaustive electrolysis experiments in stirred 1.0 M NaClO_4 solutions at 22°C using a large platinum sheet electrode ($A = 50 \text{ cm}^2$) and a constant cathodic

current of 5.00 mA. The solid line is for the solution containing added HClO_4 (0.10 ml of 0.50 M), pH = 2.55. The dashed line is for the solution containing NaCOOH (0.0089 g, 2.6 mM) and HClO_4 (0.10 ml of 0.50 M), pH = 3.34. A prior blank experiment (without any added HClO_4) reached a potential of -0.88 V within 5 seconds and contributed to higher initial pH readings.

Fig. 5. Solution pH versus time traces for the exhaustive electrolysis experiments shown in Fig. 4. The Pd-H electrode potential was measured against the SCE reference (left axis) and then converted to pH (right axis) using Eq. (1). The solid line is for the 1.0 M NaClO_4 solution containing added HClO_4 (initial pH = 2.55). The dashed line is for the NaClO_4 solution containing NaCOOH (0.0089 g) and HClO_4 (initial pH = 3.34).

Fig. 1

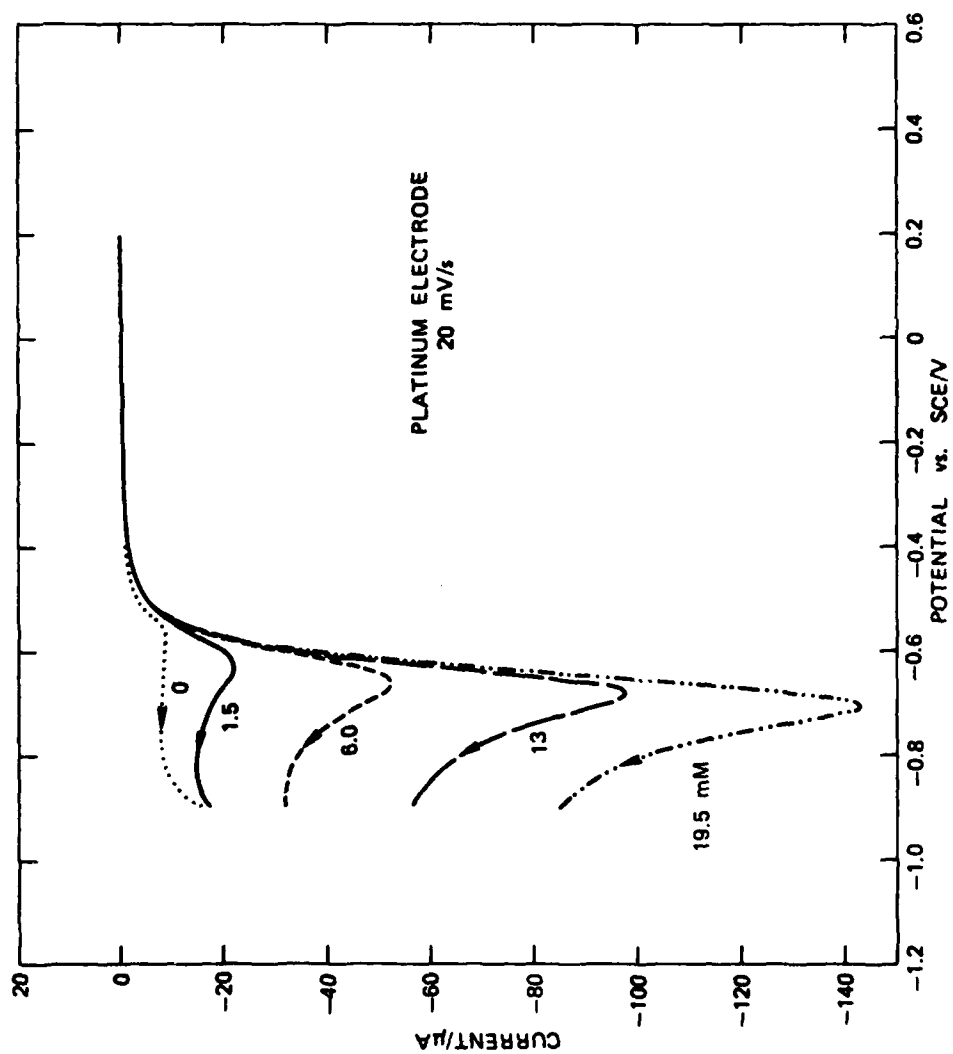


Fig. 2

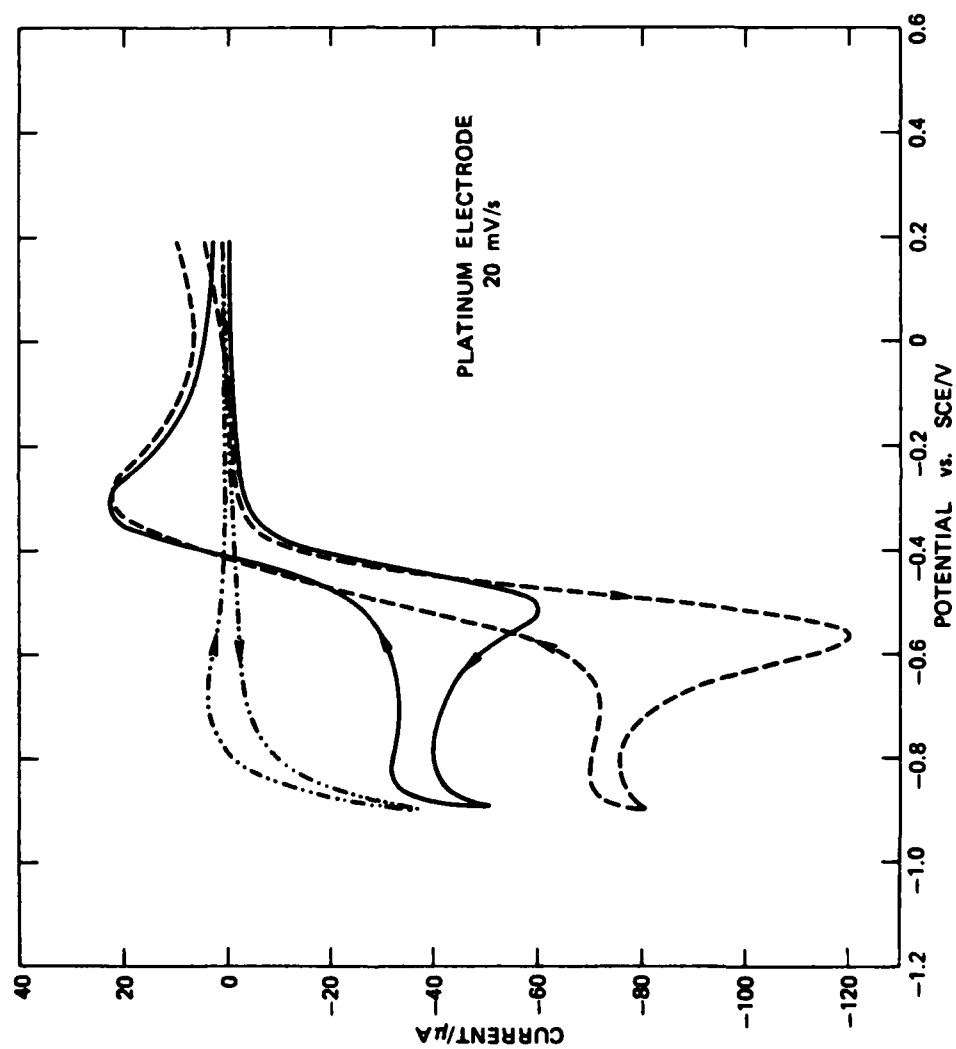


Fig. 3

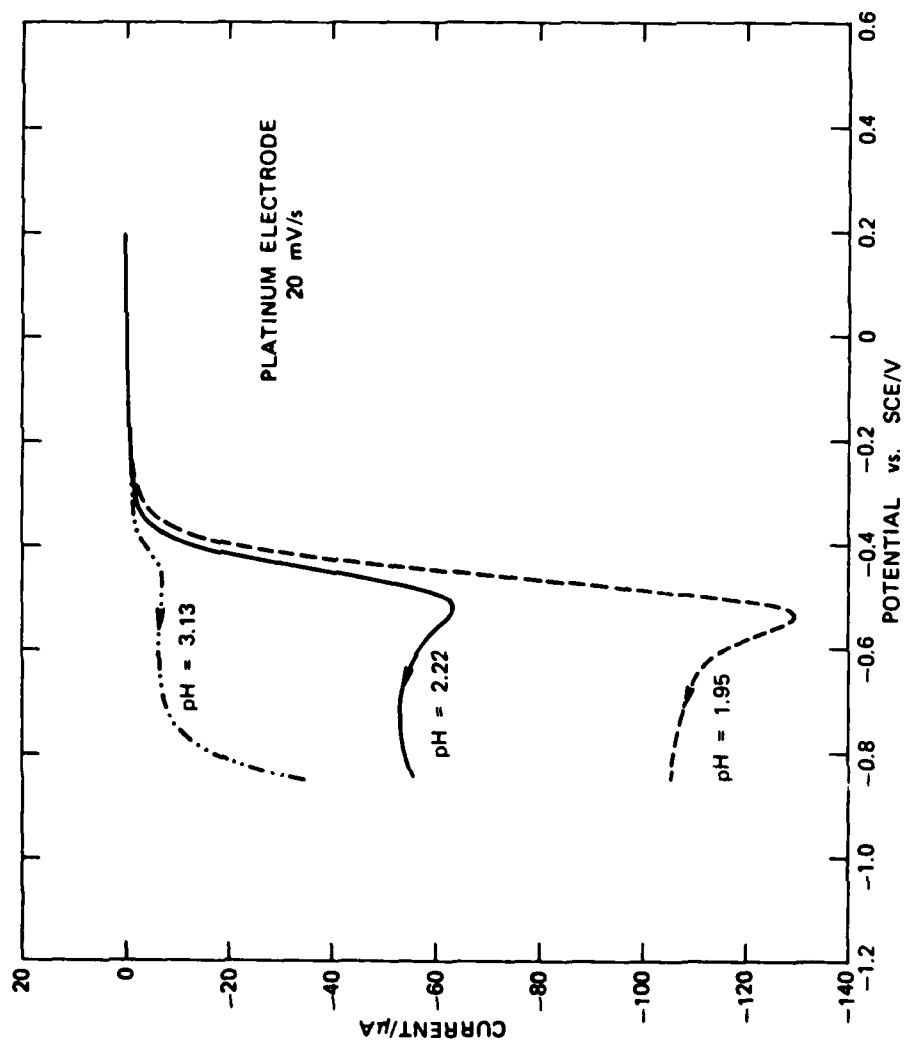


Fig. 4

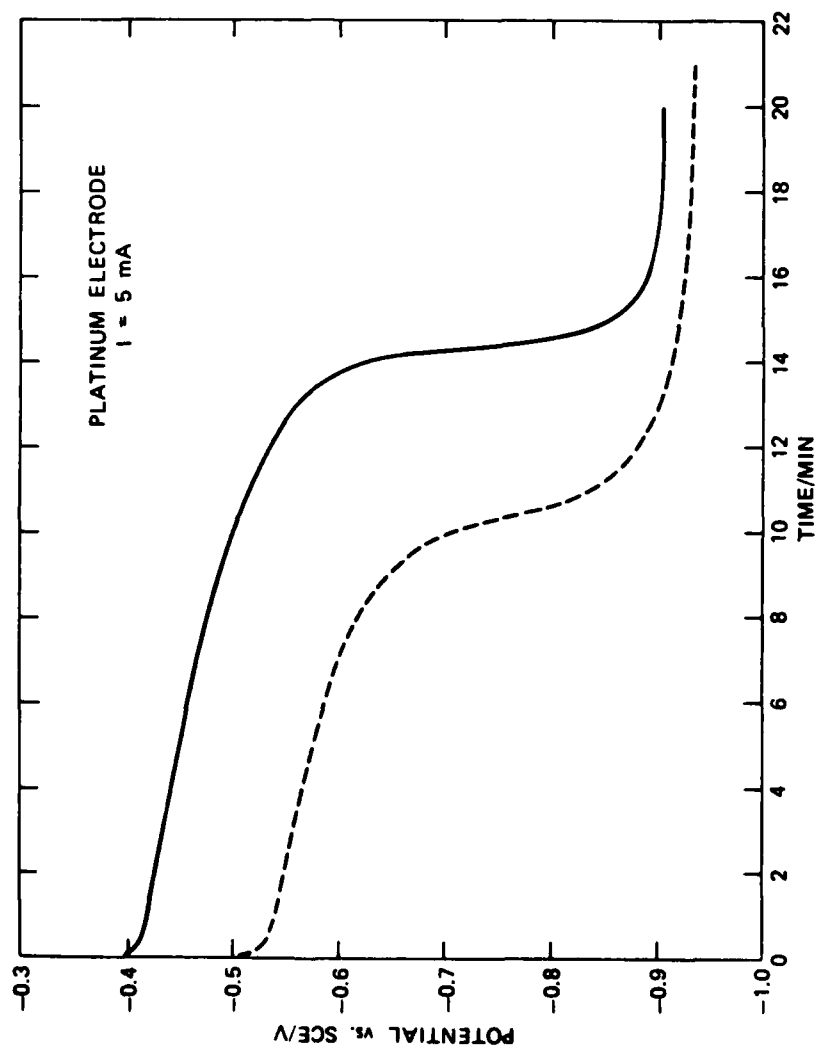
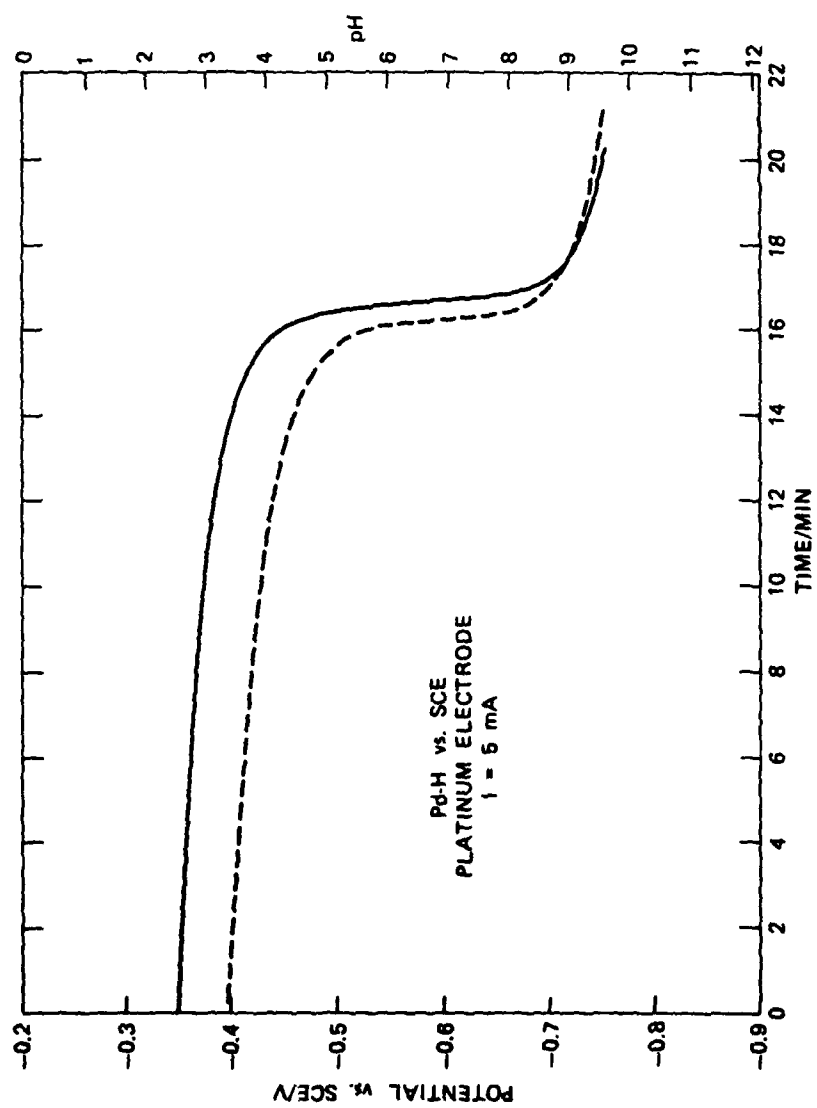


Fig. 5



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